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ATOMIC AND MOLECULAR PROCESSES IN ATMOSPHERIC
ENVIRONMENTS

Pittsburgh University

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TECHNICAL PROGRESS REPORT NO. 10

Pittsburgh Atmospheric Sciences Institute

Physics and Chemistry Departments

University of Pittsburgh

Pittsburgh, Pennsylvania

I. Summary of Research

The following research projects are identified by title, senior investigator(s) in charge of the work, and task designation(s) in the ARPA work statement. This summary only covers advances during the period since our last progress report (dated October 1, 1970).

A. Laboratory Studies

1a. Radiative Properties of Simple Molecules for Optical Discrimination Studies (F. Kaufman) (Tasks a and d).

^{to P. 2}
In the new, spherical fluorescence cell several series of experiments were carried out in order to resolve the question of the variation in the radiative lifetime of NO_2 with excitation energy which had been reported by other investigators but had not been observed by us. The excitation band width was reduced from about 50 \AA to 5 \AA , and the weak fluorescent signal was then observed through broad Corning glass filters rather than through fairly monochromatic interference filters. Six excitation wavelengths between 4050 and 4850 \AA were chosen where variations in the lifetime had been reported. No such variation was seen at NO_2 pressures near 1 millitorr where the contribution of collisional quenching is small ($\approx 20\%$) and constant. Attempts to observe the weak phosphorescence due to the low-lying triplet state of SO_2 after excitation at 3000 \AA

were unsuccessful. The radiative lifetime of the first excited singlet state (1B_1) of SO_2 was unsuccessfully measured to be 44 ± 3 μ sec by phase fluorimetry with modulated excitation. Steady excitation experiments were also carried out at various fluorescent wavelengths to ascertain whether vibrational cascading is important in the excited state. The quenching constant was indeed found to decrease with increasing $\Delta\nu = \nu_E - \nu_F$, the energy difference between excitation and fluorescent quanta, but the effect is far less pronounced in SO_2 than in NO_2 . Vibrational as well as electronic quenching in the 1B_1 state is very efficient, i.e. nearly gas - kinetic.

1b. Radiative Properties of Simple Atoms and Molecules for Optical Discrimination Studies (E. C. Zipf) (Tasks a and d).

J P 6

Substantial progress has been made during the current report period on a number of electron scattering experiments. Several papers describing this work have been submitted to the Physical Review, the Journal of Geophysics Research, and the Journal of Chemical Physics. Additional papers are in preparation. Some of our results will also be presented in three papers at the Washington meeting of the American Geophysical Union. Brief summaries of this work are presented below.

- (a) Excitation of Vacuum Ultraviolet Emission Features by Dissociative Excitation of Molecular Gases: I. H_2 and O_2 :
 The $HI(2p^1 2P - 1s^1 2S)$ 1215.67 Å Lyman alpha line and the $OI(3s^1 3S - 2p^4 3P)$ 1302.17, 1304.87, 1306.04 Å multiplet have been excited by electrons impacting on H_2 and O_2 , respectively. Their excitation cross sections have been measured over the energy range from threshold to 350 eV. The cross section for dissociative excitation of

Lyman alpha has a peak value of $1.23 \times 10^{-17} \text{ cm}^2 \pm 13\%$ while the cross section for the OI 1300 \AA multiplet has a maximum value of $3.8 \times 10^{-18} \text{ cm}^2 \pm 17\%$. The cross section for dissociative excitation of the OI($3s^1 1p^0 - 2p^4 1s$) 1217.6 \AA line is $1.5 \times 10^{-19} \text{ cm}^2 \pm 24\%$ at 100 eV.

- (b) Excitation of the CO Fourth Positive Band System by Electron Impact on Carbon Monoxide and Carbon Dioxide: Absolute excitation cross sections were measured for five vibrational bands of the CO($A^1\pi - X^1\Sigma^+$) Fourth Positive band system. The bands were produced by electron impact on CO and CO₂ and the cross sections were measured from threshold to 350 eV. Relative intensity measurements on 28 bands were used to determine the dependence of R_e on the r-centroid, which is $R_e = 1.9(1.0 - 0.6 \bar{r}_{v',v''})$. Absolute transition probabilities were computed using this functional form for R_e and normalizing the total transition probability of the $v' = 2$ level to published experimental results. The total f-value for this transition is then 0.15. Absolute excitation cross sections for the first five vibrational levels ($v' = 0 \rightarrow 4$) of the $A^1\pi$ state were determined for each parent gas using the measured cross sections of specific bands and the absolute transition probabilities. When CO was the parent gas, the relative populations of these vibrational levels were proportional to the Franck-Condon factors, $q_{v',0}$. The sum of the cross sections for these levels is $1.87 \times 10^{-17} \text{ cm}^2 \pm 15\%$ at 300 eV. This

represents 87.4% of the total excitation cross section for the $A^1\pi$ state. The relative populations of the first five vibrational levels were also determined for dissociative excitation of CO_2 and they differed from the relative populations which resulted from direct excitation of CO. The sum of the cross sections for these five levels is $1.4 \times 10^{-18} \text{ cm}^2 \pm 17\%$ at 300 eV.

- (c) Electron-Impact Excitation of the 5S State of Atomic Oxygen: The excitation of the $\text{OI}(3s^5S)$ state by electron impact on atomic oxygen has been studied in a laboratory experiment over the aeronomically important energy range from threshold to 150 eV. The $\text{OI}(^5S)$ excitation cross section was found to have shape characteristic of an optically forbidden transition involving a change of spin multiplicity and to reach its maximum value at approximately 15 eV. The absolute magnitude of the $\text{OI}(^5S)$ cross section has been estimated on the basis of laboratory data and from dayglow measurements of the $1356 \text{ \AA}/1304 \text{ \AA}$ intensity ratio. We find that $Q_{\text{max}} \approx 1.8 \times 10^{-17} \text{ cm}^2$. These studies show that photoelectron impact on atomic oxygen is the principal source of $\text{OI}(^5S)$ atoms in the dayglow. Dissociative excitation of O_2 is found to contribute less than 5% to the total 1356 \AA zenith intensity in the day airglow. The radiative lifetime of the 5S state is discussed in the light of laboratory and airglow data which suggests that actual lifetime may be shorter than the theoretical value.

- (d) **Excitation of the $\text{OI}(^3\text{S})$ and $\text{NI}(^4\text{P})$ Resonance States by Electron Impact on O and N:** The absolute cross sections for the excitation of the $\text{OI}(^3\text{S})$ and $\text{NI}(^4\text{P})$ resonance states by electron impact on atomic oxygen and nitrogen have been measured over the aeronomically important energy range from threshold to 150 eV. The peak excitation cross sections for these states were found to be $1.2 \times 10^{-16} \text{ cm}^2$ and $6.2 \times 10^{-16} \text{ cm}^2$ respectively. At low energies the total $\text{OI}(^3\text{S})$ and $\text{NI}(^4\text{P})$ cross sections exhibited well-developed peaks near 15 eV and 25 eV respectively indicating that cascade processes play an important role in exciting the $\text{OI}(^3\text{S})$ and $\text{NI}(^4\text{P})$ states.
- (e) **Photoelectron Excitation of Atomic Oxygen Resonance Radiation in the Terrestrial Airglow:** Recent laboratory measurements of the absolute cross sections for the excitation of the $\text{OI}(^3\text{S})$ resonance state by electron impact on O and O_2 when combined with in situ measurements of the photoelectron energy distribution from 120 to 300 km show that photoelectron impact is the principal excitation mechanism for $\lambda 1304 \text{ \AA}$ resonance radiation in the dayglow; dissociative excitation of O_2 is found to play a minor role. The laboratory measurements indicate that the ^3S state is strongly populated by cascade processes and imply that the $\text{OI}(3\text{p}^3\text{P} \rightarrow 3\text{s}^3\text{S}; \lambda 8446 \text{ \AA})$ transition should be a prominent dayglow emission feature. These experiments also show that the excitation of atomic oxygen by low-energy electron impact cannot account for the $\lambda 1304 \text{ \AA}$ or $\lambda 1356 \text{ \AA}$ emission observed

in the tropical ultraviolet airglow.

2. Clean Air Chemistry and Chemiluminescence (F. Kaufman) (Tasks a and d).

(a) The experimental work on the low pressure $O + NO$ chemiluminescence was virtually completed and extensive calibrations were carried out, using a standard tungsten ribbon lamp, in order to put the quantitative spectral information, obtained with six interference filters, on an absolute basis. Among the major experimental results are the following: The expected, small blue shift of the emission spectrum with decreasing pressure was observed; the fall-off in the effective second-order rate constant for chemiluminescence with decreasing pressure amounts to about a factor of six between 100 and 1 millitorr. The spectral distribution at the high pressure limit (above 100 millitorr) agrees well with the data of Fontijn, Meyer, and Schiff below 6000 \AA , but lies somewhat above their curve between 6000 and 7800 \AA . This large body of work is now being written up as a Ph.D. dissertation and will then be published in J. Chem. Phys.

(b) The study of the temperature dependence of the $O + O_3$ reaction has now been completed over the range from 296 to $409^\circ K$ and a reliable rate constant expression has been obtained, viz. $k = 1.32 \times 10^{-11} \exp(-4.24/RT) \text{ cm}^3 \text{ sec}^{-1}$ with overall probable errors in the 10 - 15% range for k . Any interference due to the presence of $O_2(^1\Delta_g)$ was minimized by producing O-atoms by the thermal decomposition of O_3 over Nernst glowers at $1900^\circ K$ in flowing Ar + O_3 gas mixtures. This work is also completed, is being written up as a Ph.D. dissertation, and will soon be published in the literature.

3. Energy Partitioning in Metal Atom Chemi-excitation (M. A. Biondi, F. Kaufman).

Early experiments have been concluded successfully on the feasibility of measuring the kinetic energy of excited metal atoms following their produc-

tion in exothermic atom-molecule reactions by means of Fabry-Perot interferometry. The 'dilute-flame' reactions between Na and Cl_2 give rise to D-line emission of Na whose line shape was measured with a pressure scanning Fabry-Perot interferometer. The principal reactions are $\text{Na} + \text{Cl}_2 \xrightarrow{1} \text{NaCl} + \text{Cl}$, and $\text{Na}_2 + \text{Cl} \xrightarrow{2} \text{NaCl} + \text{Na}^*$ in which step 2, when it produces $\text{Na}(3^2\text{P})$, is 1.2 eV exothermic. If a substantial fraction of this excess energy appears as kinetic energy of the products, the D-lines are broadened. Under the present, preliminary conditions the lines were strongly reversed because of excessive Na concentrations, and the analysis had to be confined to the wings of the line. Nevertheless it is clear (a) that the method is entirely feasible and should be pursued actively; (b) a fairly large amount of kinetic energy (~ 0.4 eV in Na^* corresponding to about 40% going into translational energy of the products) gave the best fit to the line shape data. The experimental system has now been rebuilt and will permit greatly improved experimental measurements.

4. Neutral-Neutral Reactions in Crossed Beams (W. L. Fite) (Tasks a and d).

The experimental studies of the reaction $\text{N}_2^* + \text{Na} \rightarrow \text{N}_2 + \text{Na}^*$ have now been completed, most of the analysis done, and a paper for publication is being prepared.

During the past period, the major activity has been directed to the crossed beam study of $\text{Ba} + \text{O}_2 \rightarrow \text{BaO} + \text{O}$. While results are still very preliminary, they have been enormously encouraging. It had been expected that this experiment would be a very difficult one because of possible endothermicity problems, and confusion between BaO impurity in the Ba beam and BaO formed in the reaction. In fact, both signal amplitude and signal-to-noise ratio have been much better than anticipated. The angular spread in

lab coordinates of detected BaO suggests that the process is certainly not very endothermic if at all, and must have low (if any) activation energy, in accordance with the notion that it proceeds through a harpoon mechanism. Very crude estimates of the reaction cross section at a center of mass temperature of a few hundred degrees K place it around 10^{-16} cm^2 . We are now very optimistic that variation of the Ba oven temperature will enable seeing the difference in reactivity of the singlet groundstate Ba atom and the first excited triplet metastable level.

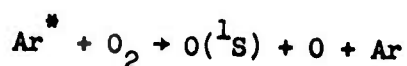
Sometimes nature really cooperates and this experiment looks like one of those times.

If what Barium seems to do with oxygen is representative of what other metals of low ionization potential do, the metal atom-oxygen molecule reactions may be a fabulous source of infra-red radiators.

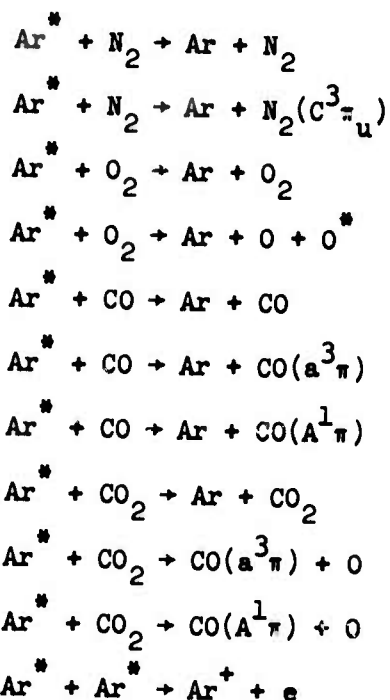
5. Processes Involving Metastable Species (E. C. Zipf) (Tasks c and d). to 8.9

Our time of flight (TOF) metastable experiments (see item 11) showed that metastable argon atoms are produced very efficiently by electron impact. It is also well known that $(\text{Ar}^* + \text{N}_2)$ energy-exchange reactions preferentially populate the $v' = 0$ and 1 levels of the $\text{C}^3\Pi_u$ state. This leads to a characteristic intensity distribution in the N_2 second positive system that differs markedly from the distribution due to electron impact. Inasmuch as argon is an important atmospheric constituent at auroral altitudes, argon metastable atoms will be produced efficiently by secondary electrons. However, the anticipated perturbation in the intensity distribution of the second positive system in an aurora has never been observed indicating that other channels more effectively deactivate the Ar^* atoms than N_2 . We have been studying this problem in some detail because it may give rise to the atomic

oxygen green line through dissociative excitation



in highly efficient manner. We have obtained data on the following collision processes:



These results are now being prepared for publication.

6. Ion Molecule Reactions and Excitation Processes in a Fast Flow System
(T. M. Donahue, W. L. Fite, and F. Kaufman) (Tasks a and b).

The study of water clusters of NO^+ and O_2^+ was continued and a large amount of kinetic information was obtained for the formation of $\text{NO}^+ \cdot \text{H}_2\text{O}$, $\text{NO}^+ \cdot 2\text{H}_2\text{O}$, $\text{NO}^+ \cdot 3\text{H}_2\text{O}$, and $\text{H}_3\text{O}^+ \cdot 2\text{H}_2\text{O}$ in the presence of He, Ar, O_2 , or N_2 as diluent carrier gases. For each of these four carrier gases six rate constants were determined by comparison of the experimentally measured ion concentrations as functions of the position of the variable water inlet tube with computer generated curves calculated for many assumed values of the rate constants. Physical consistency requires two equilibrium constants (hydration

of $\text{NO}^+ \cdot \text{H}_2\text{O}$ and of $\text{NO}^+ \cdot 2\text{H}_2\text{O}$) and one rate constant ($\text{NO}^+ \cdot 3\text{H}_2\text{O} + \text{H}_2\text{O} + \text{H}_3\text{O}^+ \cdot 2\text{H}_2\text{O} + \text{HONO}$) to be independent of the nature of M, the carrier gas. The best computer fits were found to fulfill these requirements naturally and also showed several reasonable trends: Clustering rate constants increase from $M = \text{He}$ to $M = \text{N}_2$; They increase by more than an order of magnitude for a given M as n increases from 1 to 3 in the formation of $\text{NO}^+ \cdot n \text{H}_2\text{O}$; Thermal redissociation rate constants of $\text{NO}^+ \cdot n \text{H}_2\text{O}$ increase by two orders of magnitude from $n = 2$ to $n = 3$; The "switching" rate constant from $\text{NO}^+ \cdot 3\text{H}_2\text{O}$ to $\text{H}_3\text{O}^+ \cdot 2\text{H}_2\text{O}$ is written up shortly. The O_2^+ data still suffer from some unexplained discrepancies in the $M = \text{O}_2$ or N_2 cases which cannot be properly explained by adding O_4^+ or $\text{O}_2^+ \cdot \text{N}_2$ to the reaction scheme. Further experiments are under way which should resolve this difficulty.

7. Reactions of Metal Atoms with Ions in Magnetically Confined NO^+ Plasmas
(W. L. Fite) (Tasks b and j).

During this period, the new coils that provide the magnetic confining field in the experiment have been installed. Tests on the field indicate that it is indeed much more nearly uniform than what has been used previously (which is highly desired in the interests of making analysis of experimental results less ambiguous) and the field strength in the center of the confining field approaches 1000 gauss. This latter gain is also important since keeping the Larmor radii particularly of the heavy metallic and metal oxide ions small justifies the use of adiabatic invariant approximations in the experimental analysis and again eases ambiguity problems in the interpretation of results.

The experimental work done during the period has primarily been to re-run the reactions studied in the first set-up of this experiment. The results have generally confirmed the previous results, i.e., with the alkaline earth atoms, charge transfer leaving an atomic ion strongly dominates over

ion-molecule reactions leading to polyatomic ions. Currently the data are being analyzed with a view toward arriving at good absolute cross sections for these thermal energy reactions.

The experimental progress will probably be rather slow during the next reporting period due to the need of the graduate student who has been carrying the lion's share of the work to get his thesis written.

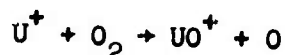
The confirming that charge transfer dominates ion-molecule reactions contains the implication that ion-electron recombination may be quite slow in heavily metal-dosed atmospheres, unless reactions of the atomic metal ions with ozone or other species operate as rapidly as some of those studied by E. E. Ferguson. It would appear desirable to have some more work done on reactions that remove atomic metal ions.

8. Ion-Molecule Reactions at Elevated Temperature (M. A. Biondi) (Tasks b and j).

Our studies of the endothermic charge transfer reaction $O^- + O_2 \rightarrow O + O_2^-$ have been completed and the results were presented by R. Johnsen at the 23rd Gaseous Electronics Conference. A characteristic feature of this reaction is the sharply rising cross section at threshold energy. The observed activation energy of ~ 1 eV is in agreement with recent photodetachment values of the electron affinities of the O_2 molecule and the O atom. The rate constant well above threshold was found to be $\sim 6 \times 10^{-11}$ cm³/sec, in agreement with data reported by other investigators. The rate coefficient exhibited a considerably sharper rise at threshold energy in our measurements than that observed in other drift tube experiments. This observation was attributed to our use of a light buffer gas (helium), rather than pure oxygen and confirms theoretical predictions that the velocity distribution of drifting ions should be narrow in light gases.

Attempts to study the reaction $H^- + O_2$ were unsuccessful, since no satisfactory ion source for H^- ions could be developed.

Recently, the drift tube has been equipped with a uranium evaporative ion source for measurements of the reactions of U^+ ions with atmospheric gases. Initial problems associated with the chemical reactivity of uranium with the tungsten heater have been solved and some data have been obtained in oxygen. These preliminary data indicate a rate constant of $(8 \pm 2) \times 10^{-10}$ cm^3/sec at $300^\circ K$ for the reaction



9. Internal Energies of Photodissociation Products (W. L. Fite) (Tasks a and d).

This experiment has been discontinued at Pitt. We understand that Dr. William Jackson of NASA Goddard Space Flight Center, who participated in the experiment during his year's visit at Pitt, may continue the experiment at Goddard.

10. Afterglow Studies of the Excitation of Radiating States by Vibrational Energy (E. C. Zipf) (Task d).

We continue with the construction of an apparatus for studying the reactions of vibrationally excited nitrogen produced by low energy electrons. This experiment essentially duplicates the conditions encountered near 105 km in an auroral arc and may contribute to our understanding of the large NO abundance observed by Zipf, Borst and Donahue. Preliminary measurements of various excitation cross sections due to electrons impacting on vibrationally excited N_2 and O_2 have been obtained.

11. Time of Flight Experiments (E. C. Zipf) (Tasks b and d).

We have been measuring the velocity of atoms produced by the dissociation of molecular targets by electron impact. These experiments use time of flight (TOF) and coincidence techniques and surface detectors to study the

metastable atoms produced by this mechanism. Our initial studies have shown that the total cross section for dissociative excitation is very large and that in many cases at least one of the atomic fragments is in a metastable state. Our time of flight apparatus has been used to study the production of $\text{CO}(a^3\pi)$ and other metastable fragments by electron impact dissociation of CO_2 . A paper describing this work has been submitted to the Journal of Geophysics Research. The principal results are the following: The dissociative excitation of $\text{CO}(a^3\pi)$ and other metastable fragments such as $\text{O}(^5\text{S}^0)$ produced by electron impact on CO_2 has been investigated from threshold to 50 eV. The observed threshold for $\text{CO}(a^3\pi)$ production at (11.9 ± 0.5) eV was near the minimum required energy of 11.5 eV. Assuming an isotropic distribution of $\text{CO}(a^3\pi)$ fragments after dissociation, the cross section for dissociative excitation of $\text{CO}(a^3\pi)$ from CO_2 was found to be about $3.6 \times 10^{-17} \text{ cm}^2$ at 20 eV and was a factor of 3 smaller than the maximum cross section for the direct excitation of $\text{CO}(a^3\pi)$ from CO. We also used our time of flight data to infer a value for the $\text{CO}(a^3\pi)$ dissociative excitation cross section from Ajello's relative cross section measurements (1971 A) and we obtained a slightly lower value of $1.6 \times 10^{-17} \text{ cm}^2$. The differential cross section for the dissociative excitation of $\text{CO}(a^3\pi)$ at right angles to the electron beam was found to be $d\sigma/d\Omega = (3.0 \pm 2.2) \times 10^{-18} \text{ cm}^2/\text{sterad}$ at an energy of 20 eV.

We have also measured the absolute cross section for the production of $\text{O}(^5\text{S})$ atoms by electron impact dissociation of O_2 . This work is described in a paper that we have submitted to the Journal of Chemical Physics. Briefly, we have combined optical observations of $\text{O}(^5\text{S})$ atoms produced by dissociative excitation with (TOF) measurements of the $\text{O}(^5\text{S})$ velocity distri-

bution. Both measurements were made as a function of the energy of the incident electron and our analysis shows that the excitation cross section has a maximum value of $1.1 \times 10^{-17} \text{ cm}^2$ near 100 eV. The geophysical implications of this result are discussed in two papers that have been submitted to the Journal of Geophysics Research.

We have also studied the production of long-lived Rydberg states of atomic oxygen and atomic nitrogen by electron impact dissociation of N_2 and O_2 . These results will be presented at the Seventh International Conference on the Physics of Electronic and Atomic Collisions. These experiments have led to a clearer understanding of the mechanism of dissociative excitation and show the important role played by atomic Rydberg states as precursors to the vacuum ultraviolet radiation observed in aurora and other atmospheric events where dissociative excitation is important.

We have made further advances in our study of the energy spectra of metastable oxygen atoms produced by electron impact. This work is described in detail in a paper accepted for publication in the Physical Review. We summarize these results briefly here: Kinetic energies of metastable O atoms formed in dissociative excitation of O_2 were measured in a time of flight experiment. Metastable $\text{O}(^5\text{S}^0)$ atoms (9.14 eV) as well as high lying states near the ionization limit of O (13.6 eV) were detected by means of Auger ejection of secondary electrons from a Cu-Be surface. The energy spectra of metastable fragments showed distinct structure with maxima near 0.3 and 2 eV. Repulsive potential energy curves of O_2 were constructed from the measured energy spectra and time-resolved excitation functions. The excitation function for the direct excitation of the $\text{O}(^5\text{S}^0)$ state had a threshold of (14.3 ± 0.2) eV which is equal to the minimum energy for dissociatively exciting the

$O(^5S^0)$ state. Using selection rules and symmetry arguments concerning the angular variation of dissociation products, the molecular state reached near 14.3 eV was identified as a π_u state with a probable multiplicity of 3. Other molecular states reached at higher energies were not identified. It seems, however, that there was a sizeable contribution due to O atoms in high lying Rydberg states excited in dissociative excitation. The differential cross section for the direct formation of $O(^5S^0)$ was found to be $d\sigma/d\Omega = 1 \times 10^{-19} \text{ cm}^2/\text{sterad}$ within a factor of two at an electron energy of 30 eV and an angle of 90° with respect to the electron beam. Several atmospheric applications of the measurements concerning the heating of the atmosphere, radiative transfer of resonance radiation and chemical reactions are mentioned briefly.

B. Ground Based Observations, Rocket and Satellite Measurements, and Aeronomy Calculations.

12. Optical Interferometer Studies of Barium Releases (M. A. Biondi) (Task g).

The telescope augmented, 100 mm Fabry Perot interferometer was used successfully to determine spectral line profiles of sunlight scattered by neutral and ionized barium in a series of releases from Eglin, Florida in January. In addition to the usual wavelength scans of the line profiles of Ba ($\lambda 5535$) and Ba^+ ($\lambda 4934$) the interferometer operated properly in a wavelength-locked mode at the hyperfine peak of the $\lambda 4934$ line. The data are being analyzed to determine the Ba cloud ionization history for comparison with the 1969 Alaska release data.

13. Optical Photometer Studies of Nightglow Ionospheric Modifications. (M. A. Biondi) (Task g).

A programmed search, filter photometer instrument which detects the spatial distribution of the atomic oxygen $\lambda 6300(^1D - ^3P)$ and $\lambda 5577(^1S - ^1D)$

intensities in twilight and nightglow has been used to observe changes in the energy of the electrons in the F region of the ionosphere induced by RF heating from a powerful (~ 1 Mw) continuous wave transmitter which is cyclically turned on and off with a ~ 10 min. period. Both intensity suppression of $\lambda 6300$ by reduction in the rate of $O_2^+ + e$ dissociative recombination and intensity enhancement by $e + O$ collisional excitation have been observed. Changes in the intensity buildup and decay time constants are being related to the (1D) quenching rates in the ionosphere--the first controlled measurement of quenching in situ.

14. Measurement of the Ionized and Neutral Constituents of the Atmosphere (T. M. Donahue and E. C. Zipf) (Task h).

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On the 25th of January (1971) we successfully launched an Aerobee 170 rocket (13.46 UA) into the dawn ionosphere above White Sands, New Mexico. The launch time was chosen so that the rocket would reach 190 km at precisely the same time that extreme ultraviolet radiation from the sun was just beginning to form the daytime ionosphere at this altitude. This was an attempt to study the transient behavior of the ionosphere as it forms and it is the first of two experiments of this kind. A second rocket (an Aerobee 350) will be launched into the twilight ionosphere above Wallops Island, Virginia during May of this year. The instrumentation on board the Aerobee 170 included a quadrupole mass spectrometer that measured the local ion and neutral composition of the upper atmosphere, two hemispherical analyzers for measuring the photoelectron energy distribution in the dawn ionosphere, a cylindrical probe for electron temperature and density measurements and a planar total positive ion probe. These particle experiments were complemented by a 1/4-meter Ebert monochromator that scanned the VUV dayglow spectrum from 1100A to 1500A and six filtered photometers that measured the overhead intensity of

the (0,0) first negative band of N_2^+ , the (5,2) first positive band of N_2 , the (0,0) second positive band of N_2 , the (1, 10) band of the Vegard-Kaplan system, and the atomic oxygen red and green lines (6300 A and 5577 A, respectively). The 6300 A and 5577 A photometers employed tilting filters to minimize the effects of scattered light.

This was basically a re-flight of the payload from Aerobee rocket 4.309 UA which encountered remarkably large concentrations of NO and NO^+ in an IBC 2^+ auroral arc. The same double-mode mass spectrometer was flown on both flights so that the White Sands experiment provided a very important calibration test for this instrument. The mass spectrometer performed perfectly in both modes during the flight of 13.46 UA and established beyond all reasonable doubt that the Churchill observations are correct (although they are certainly very difficult to explain). No nitric oxide was observed in the atmosphere above White Sands and the positive ion composition looked normal with O^+ and O_2^+ dominating the mass spectrum.

The atomic oxygen red and green line experiment was particularly successful. This involved the simultaneous measurement of the respective volume emission rates, the O_2^+ and electron density, the electron temperature, the photoelectron energy distribution, and the O, O_2 and N_2 concentrations. A preliminary analysis of the flight data has confirmed the important role played by dissociative recombination of O_2^+ ions in exciting these emission features and has yielded an $O(^1D)/O(^1S)$ branching ratio in good agreement with the laboratory results of Zipf. The measurements suggest that the branching ratio is practically independent of the electron temperature.

15. Distribution of Gases in the Mesosphere (T. M. Donahue and E. C. Zipf) (Task f).

An analysis has been made of the OGO 6 photometer data concerning

the intensity of the OI $\lambda 5577$ line emitted from the ionosphere. A model involving production of the $\lambda 5577$ radiation by dissociative recombination of O_2^+ ions and electrons in the ionosphere has been used to predict the absolute intensities observed. The efficiency of green line production by recombination deduced from the OGO 6 data is in remarkably good agreement with the value obtained from laboratory measurements.

C. Theoretical Studies

16. Semi-Empirical Methods for Estimating Reaction Rates (E. Gerjuoy) (Task e).

(a) Our calculations of charged particle-neutral atom collision cross section using the Glauber approximation continue. A paper (by B. Thomas and V. Franco) reporting our work on proton-hydrogen atom elastic and inelastic scattering has been submitted to the Physical Review. This work has been discussed in our previous technical reports. A paper (by B. Thomas and E. Gerjuoy) describing our analytic results for the Glauber amplitudes in charged particle-hydrogen atom collisions has been accepted for publication in the August issue of the Journal of Mathematical Physics. This work also was discussed in our last technical report.

One sensitive test of the correctness of the Glauber approximation is the polarization of the radiation subsequent to excitation. The polarization depends on the relative amplitudes of the various excited magnetic quantum substates, whereas the total excitation cross section (at any fixed electron scattering angle) depends only on the sum of the squares of these excitation amplitudes (for that fixed scattering angle). Thus we have begun calculations of these polarizations, taking advantage of our aforementioned analytic formulas for the scattering amplitude, which greatly simplify the numerical computations. An important feature of these Glauber calculations is recog-

dition that the implied z-axis in the Glauber formula--and therefore the convenient axis of quantization--is dependent on the scattering angle, because this z-axis is postulated (by Glauber) to be along the direction perpendicular to $\underline{k}_i + \underline{k}_f$, where \underline{k}_i , \underline{k}_f are the initial and final scattering electron wave vectors. It is anticipated that these calculations soon will be completed, and the results submitted for publication, for e-H(ls) collisions. This work is being performed by Brian Thomas, Vijay Sheorey and E. Gerjuoy. Dr. Sheorey is expected to continue cooperating in this effort (without charge to this contract) in his new position at the University of Windsor.

These polarization calculations have slowed, but not stopped, our continued efforts to extend the application of the Glauber approximation to the scattering of electrons and protons by atomic systems more complicated than hydrogen. This problem still is being worked on by Brian Thomas.

(b) Since our last report, progress in the theoretical studies of ionization and excited state populations in Ba plasmas has proceeded steadily. Some not wholly concordant results from computations on two different computers using the same programs and data forced a reexamination of the previous oscillator strength calculations. The discrepancies proved to be the result of high sensitivity in the programs; thus these calculations now are being redone in double precision. Definitive radiative recombination rates, having been otherwise completed, also await these recomputations.

The master program utilizing the various rates and other parameters to calculate the populations has been successfully debugged and tested. This work is being done by Philip Buchwalter as a Ph.D. problem; Vijay Sheorey cooperated during his tenure here.

(c) Our work under paragraph (b) above has involved use of the quantum

defect method to estimate some of the needed cross sections. Recently we have completed some other calculations using the quantum defect method. In particular, elastic phase shifts for scattering of electrons by hydrogenic positive ions have been used to calculate quantum defects--and thus energy levels--for two electron systems. Our results are in good agreement with those obtained from Z-expansion perturbation methods, Z being the nuclear charge of the system. This work performed by V. J. Sheorey, has been submitted for publication.

(d) During the past six months we have undertaken calculations of electron impact excitation cross sections for Mg^+ , Ca^+ and Ba^+ targets. These calculations were motivated by the fact that absolute measurements for electron impact excitation cross-sections of Ba^+ recently have been reported (M. O. Pace and J. W. Hooper Report No. ORO-3027-17, Contract No. AT (40-1)-3027 U. S. Atomic Energy Commission. Oak Ridge, Tennessee); Moreover, similar measurements for Ca^+ are being performed by Dunn and Taylor, at the Joint Institute of Laboratory Astrophysics, Boulder, Colorado. Thus, it is of considerable interest to compare cross-sections calculated in the Coulomb-Born approximation with these new data. Then calculations, now completed, were done by V. J. Sheorey, in collaboration with Dr. Alan Burgess, Visiting Fellow, JILA, and have been submitted for publication. The results for Ca^+ are in excellent agreement with the still unpublished results of Dunn and Taylor. During the course of this work, general programs have been developed for calculating target atom wave functions using effective potentials; this program should be of considerable use for various related collision problems.

(e) Although our work on formal three-body scattering has not been pursued during these past months, we are pleased to report that the major

content of the lengthy report by E. Gerjuoy, "Configuration Space Three-Body Scattering Theory", has been accepted for publication by the Philosophical Transactions of the Royal Society.

(f) The results of our pseudopotential calculations on lithium were so successful that a general computer code is being written so that the method can be applied to any diatomic system with few valence electrons. The basic integral routines have been completed, and work on the complete program will begin shortly. Our initial results have been published.

(g) The effects of small potential barriers on thermal collisions of atoms are being investigated. According to quantum mechanics, the particles may tunnel through such barriers and become trapped for a considerable time. We have obtained a quantum formula for the lifetime of such compound states and have used it to check previous semi-classical calculations. We found that the semi-classical results have an average error of around 10%. We are beginning to study the role of these compound states in molecular formation. Preliminary results indicate that they are important in all the cases we have studied, namely H_2 , CH and the dimers of inert and atmospheric species.

(h) We are beginning a program for calculating cross sections for electron scattering by atoms using a variational approach. Initially, we are studying resonant scattering phenomena since some experimental data of high accuracy is available for comparison. The $2S$ resonance in He^- will be the major test for our calculations. One important process for which experimental data is not available is dielectronic recombination. Experiments are planned using highly charged ions produced by passing an ion beam from a Van der Graaff accelerator through a thin foil. We have calculated the cross section for this process for O^{7+} ions. This will provide the experimenters with calibra-

tion points for these experiments which are exciting but difficult. This could be very useful because this experimental technique will make possible the accumulation of data on ions which are not easily produced by other laboratory techniques.

II. Publications and Technical Presentations

A. Publications

"Dissociative Recombination", J. N. Bardsley and M. A. Biondi, *Advances in Atomic and Molecular Physics*, 6, 1 (1970).

"Temperature Dependence of Dissociative Recombination", J. N. Bardsley, *Phys. Rev.*, A2, 1359 (1970).

"Pseudopotential Calculations of Alkali Interactions", J. N. Bardsley, *Chem. Phys. Letters*, 1, 517 (1970).

"Potential Curves of He_2^+ and Li_2^+ ", J. N. Bardsley, *Phys. Rev.* A3, 1317 (1971).

"Reactions of Na^+ , K^+ and Ba^+ Ions with O_2 , NO , and H_2O Molecules", R. Johnsen, H. L. Brown and M. A. Biondi, *J. Chem. Phys.* (to be published).

"Optical ($\lambda 6300$) Detection of Radio Frequency Heating of Electrons in the F Region", M. A. Biondi, D. P. Sipier, and R. D. Hake, Jr., *J. Geophys. Res.* 75, 6421 (1970).

"Polarization of Lyman Alpha Radiation Produced by Direct Excitation of Hydrogen Atoms by Proton Impact", W. E. Kauppila, P. J. O. Teubner, W. L. Fite, and R. J. Girnius, *Phys. Rev.* A2, 1759 (1970).

"Polarization of Lyman Alpha Radiation Produced in Charge Transfer Collisions between Protons and the Inert Gases", P. J. O. Teubner, W. E. Kauppila, W. L. Fite and R. J. Girnius, *Phys. Rev.* A2, 1763, (1970).

"Zero Field Quantum Oscillations in Beam-Foil Excited Lyman-Alpha Radiation", L. Kurzweg, W. L. Fite and R. L. Myers (submitted for publication to *Phys. Rev. Letters*).

"Lyman Alpha Emission Induced by the Collisions of Electrons with Molecular Hydrogen", W. E. Kauppila, P. J. O. Teubner, W. L. Fite and R. J. Girnius (submitted for publication to *J. Chem. Phys.*).

- "Configuration Space Three-Body Scattering Theory", E. Gerjuoy, Philosophical Transactions of the Royal Society (England), (to be published).
- "Detailed Balancing in the Time-Dependent Impact Parameter Method", E. Gerjuoy submitted to Case Studies in Atomic Physics (England).
- "Is the Principle of Superposition Really Necessary", E. Gerjuoy, to be published in the Proceedings of the Conference on the Foundations of Quantum Mechanics, University of Western Ontario, London, Ontario, March 4-7, 1971.
- "Quantum Defects for the Helium Sequence", V. J. Sheorey, submitted to J. Phys. B: Atomic and Molecular Physics (England).
- "Electron Impact Excitation of Mg II, Ca II and Ba II", A. Burgess and V. J. Sheorey, submitted to J. Phys. B: Atomic and Molecular Physics (England).
- "Glauber Amplitudes in Charged Particle Hydrogen Atom Collisions", Brian K. Thomas and E. Gerjuoy, J. Math. Phys., (to be published).
- "Elastic and Inelastic Scattering of Protons by Hydrogen Atoms", Victor Franco and Brian K. Thomas (submitted to Phys. Rev.).
- "Gas Phase Reaction Rates of Some Positive Ions with Water at 296°K", C. J. Howard, H. W. Rundle, and F. Kaufman, J. Chem. Phys. 53, 3745 (1970).
- "Oscillator Strength of the Resonance Transitions of Ground-State N and O", C. L. Lin, D. A. Parkes, and F. Kaufman, J. Chem. Phys., 53, 3896 (1970).
- "Gas Phase Kinetics of $H + H + H_2 \rightarrow 2H_2$ ", D. O. Ham, D. W. Trainor, and F. Kaufman, J. Chem. Phys. 53, 4395 (1970).
- "Kinetics and Mechanism of NO₂ Fluorescence", L. F. Keyser, S. Z. Levine, and F. Kaufman, Bull. Am. Phys. Soc. 16, 213 (1971).
- "A Mass Spectrometer Observation of NO in an Auroral Arc", E. C. Zipf, W. L. Borst and T. M. Donahue, J. Geophys. Res. 75, 6371 (1970).
- "Calibration of Vacuum Ultraviolet Monochromators by the Molecular Branching-

Ratio Technique, M. J. Mumma and E. C. Zipf, J. Opt. Soc., 61, 83 (1971).

"The $OI(6300\text{\AA})$ and $OI(5577\text{\AA})$ Intensity Ratio in the Aurora", T. D. Parkinson and E. C. Zipf, Planet, Space Sci. 1970.

"Excitation of the CO Fourth Positive Band System by Electron Impact on Carbon Monoxide and Carbon Dioxide", M. J. Mumma, E. J. Stone and E. C. Zipf, to be published, J. Chem. Phys. 1971.

"Lifetimes of Metastable CO and N_2 Molecules", W. L. Borst and E. C. Zipf, Phys. Rev. A3, 979 (1971).

"Deactivation of $N_2 A^3\Sigma_u^+$ Molecules in the Aurora", D. E. Shemansky, E. C. Zipf and T. M. Donahue, submitted to Journal Geophysics Research, 1971.

"Absolute Cross Section for the Production of $O(^5S)$ by Electron Impact Dissociation of O_2 ", W. C. Wells, W. L. Borst and E. C. Zipf, submitted to Journal of Chemical Physics, 1971.

"Photoelectron Excitation of Atomic Oxygen Resonance Radiation in the Terrestrial Airglow", E. C. Zipf and E. J. Stone, submitted to Journal of Geophysics Research, 1971.

"Electron Impact Excitation of the 5S State of Atomic Oxygen", E. J. Stone and E. C. Zipf, submitted to Journal of Geophysics Research, 1971.

"Excitation of the $OI(^3S)$ and $NI(^4P)$ Resonance States by Electron Impact on O and N", E. J. Stone and E. C. Zipf, accepted for publication in the Physical Review, 1971.

"Production of Energetic Long-Lived High-Rydberg States of Atomic Nitrogen by Electron Impact Dissociation of N_2 ", W. C. Wells, W. L. Borst and E. C. Zipf, to be published in the proceedings of the Seventh International Conference on the Physics of Electronic and Atomic Collisions, Amsterdam, 1971.

B. Technical Presentations

"Calculation of Interatomic Potentials by the Pseudopotential Method", J. N. Bardsley, at American Physical Society Conference, Seattle, 25th November 1970.

Review paper at the Symposium on Ionospheric Modification Experiments, M. A. Biondi, American Geophysical Union Meeting, December 7-10, 1970.

"Aeronomy of CO₂ Atmospheres", T. M. Donahue, at the 5th Tucson Conference on Planetary Atmospheres.

"Transfer of Excitation Energy from N₂ to Na Atoms", W. L. Fite, Colloquium talk, Bell Telephone Laboratories, Murray Hill, New Jersey, November 12, 1970.

"Transfer of Excitation Energy from N₂ to Na Atoms", W. L. Fite, Colloquium talk, Joint Institute for Laboratory Astrophysics, Boulder, Colorado, March 9, 1971.

"Transfer of Excitation Energy from N₂ to Na Atoms", W. L. Fite, Seminar talk, University of California, San Diego, April 20, 1971.

"Quantum Defect Theory", V. J. Sheorey, seminar, Wayne State University, Detroit, Michigan, November 3, 1970.

"Is the Principle of Superposition Really Necessary?", E. Gerjuoy, invited paper, at the Conference on the Foundations of Quantum Mechanics, University of Western Ontario, London, Ontario, March 4-7, 1971.

"Classical Calculations of Atomic Collision Cross Sections", E. Gerjuoy, Physics Colloquium, Lycoming College, Lycoming, Pa., November 16, 1970.

"Classical Calculations of Atomic Collision Cross Sections," E. Gerjuoy, invited paper, American Association of Physics Teacher, Western, Pa. Section, Fall Meeting, Carnegie-Mellon University, Pittsburgh, Pa., November 7, 1970.

"Configuration Space Theory of Three-Body Scattering," E. Gerjuoy, University

of Pittsburgh and Carnegie-Mellon Joint Theoretical Seminar, November 4, 1970.

"Classical and Semi-Classical Methods of Computing Atomic Collision Cross Sections", E. Gerjuoy, Colloquium, Wayne State University, October 21, 1970.

"Classical and Semi-Classical Methods of Computing Atomic Collision Cross Sections", E. Gerjuoy, colloquium, City University of New York, November 11, 1970.

"Proton-Hydrogen Scattering", contributed paper by Victor Franco and Brian Thomas (presented by E. Gerjuoy) to the American Physical Society Division of Electronic and Atomic Physics Meeting, Seattle, Washington, November 23, 1970.

"Glauber Amplitudes in Electron Hydrogen Collisions", E. Gerjuoy and B. Thomas, American Physical Society Division of Electronic and Atomic Physics Meeting, Seattle, Washington, November 24, 1970.

"Rates of Formation of Water Clusters for O_2^+ and NO^+ ", C. J. Howard, H. W. Rundle, and F. Kaufman, paper presented at the 23rd Gaseous Electronics Conference, Hartford, Conn., October 23, 1970.

"Thermal Ion-Molecule Reactions Involving Water", F. Kaufman, Bushy Run Radiation Laboratory of Mellon Institute, Seminar, October 28, 1970.

"Thermal Ion-Molecule Reactions Involving Water", F. Kaufman, Wayne State University, seminar, November 11, 1970.

"Reactions of Some Minor Constituents in CO_2 Atmospheres", F. Kaufman, invited review paper at Fifth Arizona Conference on Planetary Atmospheres, Kitt Peak National Observatory, Tucson, Arizona, March 8, 1971.

"Some Implications of a Large NO Abundance in an Auroral Arc", E. C. Zipf, W. L. Borst and T. M. Donahue, Trans. AGU., 51, 790 (1970). Paper presented at San Francisco meeting of the American Geophysical Union, December 1970.

"Deactivation of $N_2 A^3 \Sigma_u^+$ Molecules in the Aurora", T. M. Donahue, E. C. Zipf and D. E. Shemansky, Trans. AGU. 52, 309, (1971). Paper presented at Washington meeting of the American Geophysical Union, April 1971.

"Differential Electron Flux as Determined by Auroral Observations of the N_2 Positive and N_2^+ Systems", D. E. Shemansky, T. M. Donahue, and E. C. Zipf, Trans. AGU, 52, 305 (1971). Paper presented at Washington meeting of the American Geophysical Union, April 1971.

"Excitation of the $OI(^3S)$ and $OI(^5S)$ States by Electron Impact on O and O_2 in the Aurora, Dayglow and EEUV", E. C. Zipf, E. J. Stone, M. J. Mumma, W. C. Wells, and W. L. Borst, Trans. AGU. 52, 308 (1971). Paper presented at Washington meeting of the American Geophysical Union, April 1971.

C. Other Activities.

J. N. Bardsley has made several visits to Goddard Space Flight Center, Washington, for consultation with Dr. Temkin's group.

R. D. Hake, D. P. Sipler, and M. A. Biondi participated in the barium release series at Eglin, Florida in January and February 1971, carrying out observations of the spectral line profiles of Ba and Ba^+ .

R. D. Hake and M. A. Biondi attended the preliminary data reduction meeting concerning the Florida barium releases at S.R.I. March 30-31, 1971.

M. A. Biondi spent three weeks at the Center for Theoretical Studies at the University of Miami in February 1971 writing an Essay entitled "Atomic Collision Physics--Its Influence on Technology and Society" for the Series "The Impact of Basic Research on Technology."

E. Gerjuov participated in the regular annual meeting of the Oak Ridge National Laboratory Health Physics Visiting Committee, Oak Ridge, Tennessee, October 27-30, 1970.

- E. Gerjuoy attended the meeting of the American Physical Society Division of Electronic and Atomic Physics, Seattle, Washington, November 23-25, 1970.
- E. Gerjuoy attended the Conference on Relativistic Astrophysics, University of Texas, Austin, Texas, December 14-18, 1970.
- E. Gerjuoy, at the invitation of the Department of Physics, Texas A & M University, spent December 10-12 in College Station, Texas, discussing atomic and molecular collision theory.
- E. Gerjuoy attended the IDA, Washington D. C., "Reentry and Atomic Physics Meeting", called by ARPA, November 5, 1970, where he reported on the PASI Theoretical Program.
- F. Kaufman attended an ARPA sponsored meeting on Reentry Atomic and Molecular Physics and presented a review of part of PASI experimental work, Washington, D. C., October 5 and 6, 1970.
- F. Kaufman attended a DASA meeting on computer codes in Washington, D. C., December 10, 1970.
- F. Kaufman attended a meeting of the DASA Reaction Rate Working Group in Washington, D. C., January 12, 1971.
- F. Kaufman accepted an invitation to be a member of a panel of four to plan the overall program of the 14th International Combustion Symposium and attended a panel meeting at Linden, N. J., February 25, 1971.
- F. Kaufman was invited to be an advisor to the SST Environmental Research Panel of the U. S. Department of Commerce Technical Advisory Board and attended a panel meeting in Boulder, Colorado, March 18 and 19, 1971.
- E. C. Zipf launched a Nike-Apache rocket and an Aerobee 170 rocket from White Sands, New Mexico on November 25, 1970 and January 25, 1971 respectively to study the day airglow and the ion and neutral composition of the mid-latitude atmosphere.

E. C. Zipf is now involved in a joint biophysics experiment with the Department of Microbiology of the University of Maryland which exploits many of the signal averaging techniques developed by his group to study the behavior of pathogenic and non-pathogenic bacteria.

IV. Visiting Scientists

Dr. Stephen Ormonde, Quantum Systems, Inc.

H. Michaels, United Aircraft

Robert Mace, Army Research Office - Durham

Frank P. Del Greco, Air Force Cambridge Research Laboratories

Peter Siska, University of Chicago

Dudley R. Herschbach, Harvard University

Ralph Kummner, Wayne State University

Soji Tsuchiya, Cornell University

Paul Feldman, Johns Hopkins University

V. Degrees Awarded

Herbert F. Krause, Ph.D., October 1970.

Lutz Kurzweg, Ph.D., October 1970.

<u>Senior Investigator</u>	<u>Est. Funds Expended and Committed (Thousands)</u>
J. N. Bardsley	19.2
M. A. Biondi	259.0
T. M. Donahue	234.4
W. L. Fite	252.0
E. Gerjucy	234.2
F. Kaufman	242.3
E. Zipf	233.3
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Total Expended and Committed	1,474.4
Available Funds	1,649.9
Estimated Remaining Funds as of 4/1/71	<u><u>175.5</u></u>

University Accounting of Funds

Expended as of 3/31/71	\$ 1,457.4
Available Funds	<u>1,649.9</u>
Remaining Funds as of 4/1/71	\$ 192.5
